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Levels of Lead, Cadmium, and Mercury in Canadian Cigarette Tobacco as Indicators of Environmental Change: Results from a 21-Year Study (1968–1988)

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Close to 90% of Canadian domestic cigarette tobacco is grown within a small geographic area in southern Ontario. The lead, cadmium, and mercury contents of this geographically homogeneous plant material was determined as a potential indicator of environmental change. During the 21-year period (1968–1988) studied, the major decreases in concentrations of lead, cadmium, and mercury occurred during a 7–9-year period commencing about 1972. With respect to lead, this period of major change probably reflects progress in reducing levels in ambient air. Concentrations since 1979 have either stabilized (mercury) or decreased at a much slower rate (lead and cadmium) and may indicate that residual levels in the soil are now the primary determinants of uptake into tobacco plants in southern Ontario. From 1968 to 1988 levels of lead, cadmium, and mercury in mainstream smoke, which is inhaled by smokers, are estimated to have declined by about 62%, 39%, and 52%, respectively.

Introduction

Domestic brands of Canadian cigarettes are unique in that 90% of the tobacco used in their production is flue-cured and grown in the Province of Ontario (*1*). Within Ontario, 97% of all tobacco is grown in just five counties (*2*). In contrast, cigarettes in other countries are often composed of tobacco blends originating potentially from many geographic locations (*3*). In the United States, for example, one study has characterized U.S. cigarette tobacco as being 34% flue-cured, 28% burley, 15% oriental, 2% maryland, 21% not defined, and 33% imported (*3*).

Given the unique homogeneity of Canadian cigarette tobacco, Canadian cigarettes are a unique source of data concerning changes in the levels of some environmental toxic constituents that have been taken up by the tobacco plant over time. This is particularly true of environmental contaminants, which transferred to the tobacco plant from the soil or deposited on the leaves. Trace metals such as lead, cadmium, and mercury are typical examples and are the focus of the present study. The approach used in this investigation is the same as the one employed in a study of cigarette tobacco from western Europe. In that case, a comparison of 1987 levels with 1991 levels of lead, cadmium, and mercury showed a clear reduction, which was attributed to environmental protection measures (*4*).

Lead, cadmium, and mercury are ubiquitous metals with known toxic properties (*5, 6*). Historically, the presence of lead in whole tobacco has resulted from both soil uptake and leaf deposition due to automobile emissions, a primary cause of lead pollution.

For example, in 1982, total emissions of lead in Canada were estimated at 11 466 t, with gasoline-powered motor vehicles accounting for 61% (*7*). Levels in soil reflect both ambient air concentrations and agronomic practices, such as the use of pesticides and sludge as a fertilizer (*8, 9*). Consequently, decreases in toxic trace metals in air would not necessarily result in similar changes in soil.

The cadmium content of tobacco has also been examined in a number of studies (*10, 11*) since cigarette smoke may be the most significant source of this toxic element in the smoking population (*12*). Levels in whole tobacco vary considerably from country to country, with cigarettes from Sri Lanka and Mexico containing 0.46 and 4.41 μg/g, respectively (*10*).

Mercury content of tobacco has also been of interest due to its toxicity and its historical use in mercurial fungicides formulated for tobacco usage. The mercury content of cigarette tobacco has been reported as 30 ± 10 ng/cigarette (*13*).

When considering possible health implications for smokers, it is important to note that the trace element content of tobacco serves as an upper bound for concentration in smoke. With respect to cadmium, when tobacco is smoked, the transference of inhaled constituents is 20% or less (*12*) and falls to about 6% for lead (*14*).

Materials and Methods

Cigarette Samples. Every year since 1968, with the exception of 1975, the Government of Canada has carried out one or more surveys of yields of tar, nicotine, and other tobacco smoke constituents. Representative brands of Canadian cigarettes are purchased for this purpose on the open market by members of the Health Protection Branch, Health and Welfare Canada. Upon reaching the testing facility, it has been the practice to store a sample of each of the major sellers as defined by Canadian Domestic Sales Protocol of 1968. This inventory, which is stored under conditions which minimize deterioration, has been used previously for investigations of historical trends (*15*).

In 1968, five brands accounted for approximately 30% of Canadian sales and were selected for this study (*16*). One package of each brand for each year (1968–1988) was removed from storage, giving a total of 100 brand/year combinations. Each brand/year combination was assigned a number, and analyses were carried out in random order. The experiment consisted of three replicates for a total of 300 observations for each metal.

Analytical Procedures. Ten cigarettes were taken from each package, the paper and filter were removed, and the pooled sample was placed in glass vials. A 2-g aliquot was taken from each pooled sample and lyophilized

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for 72 h, and the moisture content was estimated from the weight loss.

Samples were digested by first weighing 2 g of freeze-dried tobacco into a 200-mL reflux-type digestion tube. A 20-mL sample of environmental grade HNO_3 (nitric acid) was added, and the mixture was allowed to stand for 12 h.

Samples were digested at 60 °C for another 12 h (Labconco Rapid Digest-4), and the temperature was then increased to 90 °C for 1 additional hour. After a further 2-h digestion at 120 °C, 10 mL of 30% H_2O_2 (hydrogen peroxide) was added in small aliquots, and the sample was heated for another 2 h. The digestate was filtered, made to 100 mL using type I water, and then transferred to polyethylene bottles with Teflon caps. In order to determine background levels, the procedure was carried out without a tobacco sample (a control sample).

Lead and cadmium were determined by flame atomic absorption spectrometry using a Varian 8 lamp Spectra AA-400 Series instrument equipped with a programmable sample changer. Mercury was determined by flameless cold vapor atomic absorption spectroscopy using a Varian VGA-76 vapor generator in conjunction with the Spectra AA-400.

Recovery of lead, cadmium, and mercury from spiked tobacco indicated no appreciable loss during digestion. Levels of lead, cadmium, and mercury in blanks were negligible.

Results

Initially, the data were examined using a two-way analysis of variance in which "year" was treated as a covariate. The year effect for all three metals was highly significant, but there was no evidence for either a brand effect within a year or a difference among the three replicates.

Yearly averages taken over replicates (three) and brands (five) are shown in Figures 1 and 2. Error bars are standard deviations and are obtained from the five brand averages within each year. All values have been expressed on a dry weight basis.

Changes in the lead content of manufactured tobaccos (Figure 1) are compared with levels of lead in ambient air as recorded in ref 17. The differences in scales make a direct comparison difficult but it is clear that both decreased significantly during the time period 1974–1988. Using 1974 as the base year and taking concentrations in 1974 as 100%, a regression of percentage of 1974 levels on time was carried out. Based on the estimated slope of the least-squares regression line, levels of lead in ambient air decreased 5.4% per year (±0.17%) from 1974 to 1988, while lead in tobacco decreased by about half as much (2.6% per year ±0.33%).

A regression of tobacco lead on air lead was also carried out and resulted in a R^2 value of 90%. Consequently, for this data set, variations in ambient air lead account for most of the corresponding variation in tobacco lead. The regression analysis also demonstrated that for this data set, lead in tobacco leaf can be used to estimate lead in ambient air with a reasonable level of precision (coefficient of variation of 10.1%).

Levels of cadmium and mercury have also decreased since 1968 (Figure 2). In each case, the relationship is not simple, and several attempts were made to fit the data.

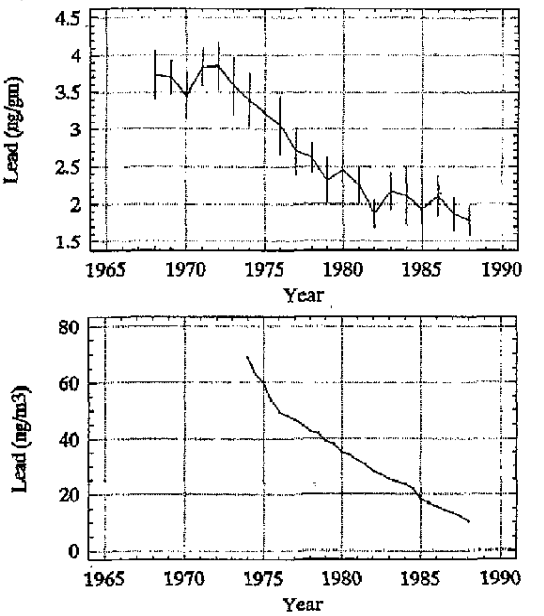

Figure 1. Comparison of the lead content of manufactured cigarette tobacco (upper section) with concentrations of lead in ambient air (lower section taken from ref 17). Error bars represent the standard deviation for the five brand averages within each year.

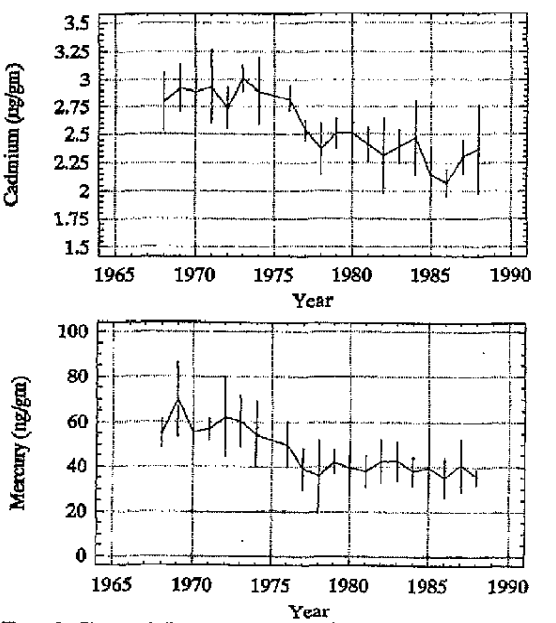

Figure 2. Changes in the average cadmium (upper section) and mercury (lower section) contents of manufactured cigarette tobaccos during the period 1968–1988 (see Figure 1 and the text for additional details).

Each model suffered from a serious and obvious lack of fit as determined from an examination of residual plots. Nevertheless, the interpretation of the data seems fairly clear. From 1968 until about 1972, levels of lead, cadmium, and mercury in whole tobacco were fairly constant,

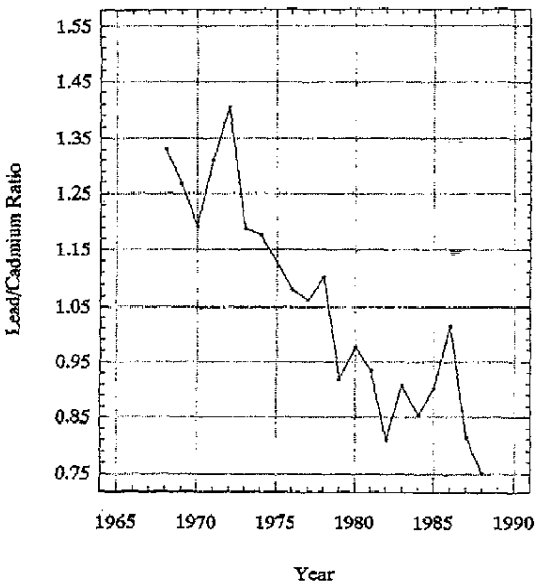

Figure 3. Changes in the average lead/cadmium ratio during the time period 1968–1988.

averaging 3.71 μg/g, 2.88 μg/g, and 60 ng/g, respectively. From about 1972 to 1975, levels of all three metals began to decrease with lead showing the largest decrease (0.178 μg g^{-1} $year^{-1}$) and mercury showing the smallest (4.89 ng g^{-1} $year^{-1}$).

Mercury concentrations stabilized at about 21 ng/g around 1977–1979 and remained at that level for the remainder of the study period. On the other hand, lead and cadmium concentrations in whole tobacco continued to decline but at lower rates beginning about the same time (1979) for cadmium and a bit later (1983) for lead.

Discussion

Toxic trace elements are present in tobacco as a result of a combination of both air deposition and soil uptake, with the physiology of the plant determining the importance of the particular source. Since lead is usually immobilized in soils, absorption by the tobacco plant does not depend directly on soil concentration (*18*). Consequently, it has been reported that the lead content of soil is unrelated to the lead content of tobacco leaves (*8*).

Cadmium, on the other hand, is preferentially taken up from the soil due to the induction of the cadmium binding protein metalothionein (*19*). The affinity of the tobacco plant for cadmium is well-illustrated by the finding that 97% of the variation in tobacco leaf cadmium was due to variations in the content of cadmium in the soil (*8*). This preferential cadmium uptake is responsible for a lead to cadmium ratio in tobacco of about 1 order of magnitude, less the ratio of 5 generally found in other plant samples (*20*). Thus, the observed steady decline in the lead cadmium ratio for Ontario tobaccos is in keeping with reductions in ambient air lead and the known physiology of the tobacco plant (see Figures 1 and 3).

Mercury is taken into the plant primarily by the growing root and subsequently translocated to the leaves along with other nutrients. In 1976, the mercury content of 112 sandy soils from Ontario ranged from 10 to 70 ng/g with a mean of 60 ng/g (*21*). Tobacco grown at that time on the same type of soil had a mean mercury content of 50.1 ng/g. In contrast, the corresponding soil (S) and tobacco (T) lead and cadmium contents were as follows: Cd-S, 0.43; Cd-T, 2.82; Pb-S, 10.4; and Pb-T, 3.05 μg/g based on the soil data from ref 21 and the results from this study. In summary, it is not unreasonable to assume that the lead content of tobacco is related primarily to ambient air concentrations while the cadmium content reflects primarily soil concentrations. Mercury content is probably a reflection of contributions from both air and soil.

Table 1. Changes in Levels of Lead, Cadmium, and Mercury in Whole Tobacco and Mainstream Tobacco Smoke

A. Whole Tobacco

av content	Time Period 1968–1971	1985–1988
lead (μg/g)	3.68 ± 0.063[a]	1.92 ± 0.060
cadmium (μg/g)	2.89 ± 0.055	2.22 ± 0.051
mercury (ng/g)	58.4 ± 1.83	37.7 ± 2.32

B. Mainstream Smoke[b]

	year 1968	1988	decrease (%)
lead (ng/cigarette)	216	83	62
cadmium (ng/cigarette)	271	164	39
mercury (ng/cigarette)	5.8	2.8	52

[a] Standard error for the mean. [b] Estimated from published values for smoke transferance, which are lead 6% (*13*); cadmium 10% (*14*); and mercury 10% (*14*), and weight of tobacco per cigarette (*23*).

In the strictest sense, the above discussion applies only to the tobacco leaf since there are many stages between the green leaf and manufactured cigarette tobacco; at each stage there is the possibility of contamination. Although this type of contamination cannot be ruled out, the consistency of the data (see Figure 1) and the agreement of levels with the data from similar studies from a number of other countries make this unlikely (*4, 10, 11, 13, 14*). It should also be noted that the tobacco in any given cigarette is a mixture coming in from many different forms. As such, it would be expected to be more reflective of toxic trace metal contamination in soil or air than any single leaf or sample of leaves from a single farm.

Lead, cadmium, and mercury are all toxic trace elements with well-documented health effects (*6*), and in each case, tobacco usage is a contributor to the total body burden. This is particularly true of cadmium where tobacco has been estimated to be the single most important nonindustrial source for exposure in smoking populations (*13*). It is reasonable to assume, then, that significant decreases in the toxic trace metal content of whole tobacco would result in lower levels in tobacco smoke and a reduction in cadmium uptake by Canadian smokers. Of course, this assumes that all other variables such as smoking behavior have remained constant during the interval (*22*).

The results, summarized in Table 1, show a significant decrease since 1968 in the potential contribution of cigarette smoking to the body burden of these elements. While a part of the change is due to a 20% reduction in the tobacco content of cigarettes (*23*), the larger part of the decrease is due to lower concentrations of heavy metals in the tobacco itself.

As other sources of these toxic chemicals are eliminated or as emissions are reduced, tobacco has become one of the major sources of lead and cadmium to a significant fraction of the population. In Canada, the smoking prevalence declined from approximately 50% in 1965 to approximately 32% in 1989 (24). Since the brands chosen for this study represented from 19% to 27% of total Canadian cigarette sales during the period 1968–1988, the public health community and environmentalists can now estimate the contribution of cigarette smoking to heavy metal exposure for a significant proportion of the population during this 20-year period.

Acknowledgments

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